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## Improved Evaluation of Electrical Phase Parameters in a Dielectric Theory for Concentrated Suspensions of Spherical Particles

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An improved method is proposed to find a physicochemically significant solution of the equation which was presented in a previous paper [*Bull. Inst. Chem. Res., Kyoto Univ.*, **55**, 376 (1977)] to evaluate electrical conductivity of the continuous phase in a concentrated suspension of spherical particles. An existing domain of the complex permittivity of the suspension was made clear in a permittivity-conductivity plane with mathematical formulas as well as with graphic representation. As a consequence, the searching scheme for the root was refined so as to attain to the correct root of the equation straightway. Some numerical examples are shown to confirm the validity of the improved method.

KEY WORDS: Concentrated suspension/ Dielectric relaxation/ Electrical conductivity/  
Interfacial polarization/ Maxwell-Wagner relaxation/ Permittivity/

### I. INTRODUCTION

It was reported by a number of workers<sup>1-3)</sup> that coarsely dispersed systems such as emulsions and suspensions showed remarkable dielectric relaxations, which were understood to be caused by the interfacial polarization due to their diphasic structure.

Such dielectric relaxations caused by interfacial polarization for suspensions of spherical particles were first pointed out by Maxwell,<sup>4)</sup> having been afterwards formulated by Wagner<sup>5)</sup> in a form convenient for the comparison with experiments. Since closer consideration revealed that Wagner's equation was in poor agreement with experiments at higher concentrations of the suspended particles, Hanai<sup>6,7)</sup> proposed a theoretical equation which is expected to be applicable up to higher concentrations.

On the basis of Hanai's equation, a systematic method<sup>8)</sup> was proposed to calculate the relative permittivity, the electrical conductivity, and the concentration of the suspended particles by use of the observed data on dielectric relaxation caused by a diphasic structure of spherical disperse systems. The method proposed, however, was not necessarily successful in evaluating the electrical conductivity of the suspended particles in such a way that the computer searching for the solution of an equation in the theory sometimes takes hold of a false root.

In the present paper, an effectual method is proposed to find a correct root of the equation. Several examples are illustrated to show merits of the proposed method on computer searching for the correct roots of the equation.

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## II EQUATIONS FOR CONCENTRATED DISPERSE SYSTEMS OF SPHERICAL PARTICLES

In order to make clear the circumstances in question, we shall summarize the equations used.<sup>8)</sup>

For a concentrated disperse system of spherical particles with a complex permittivity  $\epsilon_i^*$  in a continuous medium with a complex permittivity  $\epsilon_a^*$ , Hanai derived the following equation for the complex permittivity  $\epsilon^*$  of the whole system:

$$\frac{\epsilon^* - \epsilon_i^*}{\epsilon_a^* - \epsilon_i^*} \left( \frac{\epsilon_a^*}{\epsilon^*} \right)^{1/3} = 1 - \phi, \quad (1)$$

where  $\phi$  is the volume fraction of the dispersed particles. The complex permittivities  $\epsilon^*$ ,  $\epsilon_a^*$  and  $\epsilon_i^*$  are each defined by the following equations:

$$\epsilon^* = \epsilon - j \frac{\kappa}{2\pi f \epsilon_v}, \quad (2)$$

and

$$\epsilon_a^* = \epsilon_a - j \frac{\kappa_a}{2\pi f \epsilon_v}, \quad (3)$$

and

$$\epsilon_i^* = \epsilon_i - j \frac{\kappa_i}{2\pi f \epsilon_v}, \quad (4)$$

where  $j$  is  $\sqrt{-1}$ ,  $f$  the frequency,  $\epsilon$  the relative permittivity,  $\kappa$  the electrical conductivity and  $\epsilon_v$  the permittivity of free space. The subscripts  $a$  and  $i$  refer to the continuous medium and the suspended particles, respectively.

The limiting values of  $\epsilon$  and  $\kappa$  at high (subscript  $h$ ) and low ( $l$ ) frequencies are given by

$$\frac{\epsilon_h - \epsilon_i}{\epsilon_a - \epsilon_i} \left( \frac{\epsilon_a}{\epsilon_h} \right)^{1/3} = 1 - \phi, \quad (5)$$

$$\epsilon_l \left( \frac{3}{\kappa_l - \kappa_i} - \frac{1}{\kappa_l} \right) = 3 \left( \frac{\epsilon_a - \epsilon_i}{\kappa_a - \kappa_i} + \frac{\epsilon_i}{\kappa_l - \kappa_i} \right) - \frac{\epsilon_a}{\kappa_a}, \quad (6)$$

$$\kappa_h \left( \frac{3}{\epsilon_h - \epsilon_i} - \frac{1}{\epsilon_h} \right) = 3 \left( \frac{\kappa_a - \kappa_i}{\epsilon_a - \epsilon_i} + \frac{\kappa_i}{\epsilon_h - \epsilon_i} \right) - \frac{\kappa_a}{\epsilon_a}, \quad (7)$$

and

$$\frac{\kappa_l - \kappa_i}{\kappa_a - \kappa_i} \left( \frac{\kappa_a}{\kappa_l} \right)^{1/3} = 1 - \phi. \quad (8)$$

Provided that  $\epsilon_a$ ,  $\epsilon_h$ ,  $\epsilon_l$ ,  $\kappa_h$  and  $\kappa_l$  are given from the observed data, we can obtain the values of  $\kappa_a$  by means of computer searching for the root of the following equation which was derived from Eqs. 5-8:

$$\begin{aligned}
 J(\kappa_a) = & [3 - (2 + \frac{\varepsilon_a}{\varepsilon_h}) C] (1 - DC) \kappa_h \\
 & - 3[(1 - C) \kappa_l + (1 - D) C \kappa_a] (1 - C) \\
 & + (1 - \frac{\varepsilon_h}{\varepsilon_a}) (1 - DC) C \kappa_a = 0,
 \end{aligned} \tag{9}$$

where

$$C = \frac{-Q - \sqrt{Q^2 - 4PR}}{2P}, \tag{10}$$

$$P = (2 + \frac{\kappa_a}{\kappa_l}) D \varepsilon_l - 3[D \varepsilon_h + (1 - D) \varepsilon_a] D + (\frac{\kappa_l}{\kappa_a} - 1) D \varepsilon_a, \tag{11}$$

$$Q = 3[2D \varepsilon_h + (1 - D) \varepsilon_a] - [(2 + \frac{\kappa_a}{\kappa_l}) D + 3] \varepsilon_l - (\frac{\kappa_l}{\kappa_a} - 1) D \varepsilon_a, \tag{12}$$

$$R = 3(\varepsilon_l - \varepsilon_h), \tag{13}$$

$$D = (\frac{\varepsilon_a \kappa_l}{\varepsilon_h \kappa_a})^{1/3}. \tag{14}$$

Using the values of  $\kappa_a$  thus obtained, values of  $\phi$ ,  $\varepsilon_i$  and  $\kappa_i$  are calculated from the following equations in sequence:

$$\phi = 1 - C(\frac{\varepsilon_a}{\varepsilon_h})^{1/3}, \tag{15}$$

$$\varepsilon_i = \frac{\varepsilon_h - C \varepsilon_a}{1 - C}, \tag{16}$$

and

$$\kappa_i = \frac{\kappa_l - DC \kappa_a}{1 - DC}. \tag{17}$$

According to the remarks shown in Appendices of the previous paper,<sup>8)</sup> the computer searching for the root of Eq. 9 should be proceeded from  $\kappa_l$  toward the lower values of  $\kappa_a$  when  $\kappa_a < \kappa_l$ . If  $\kappa_l < \kappa_a$ , then the searching should be proceeded from  $\kappa_l$  toward the higher values of  $\kappa_a$ . After several trials of numerical calculations, it turned out incomplete to perform the above computer searching.

An improved computer searching is proposed on the basis of the discussion developed in the succeeding sections.

### III. GRAPHIC EXPLANATION CONCERNING THE LOCATION OF $\varepsilon^*$ ON THE $\varepsilon$ - $\kappa$ PLANE

For the purpose of discussing the dielectric relaxation caused by the diphasic structure, the representation of  $\varepsilon^*$  (abscissa  $\varepsilon$ , ordinate  $\kappa$ ) on an  $\varepsilon$ - $\kappa$  plane is more comprehensible and convenient as compared with the conventional plots on  $\varepsilon$ - $\varepsilon''$  planes.

In dielectric relaxation phenomena, the relative permittivity  $\varepsilon(f)$  is a monotonically



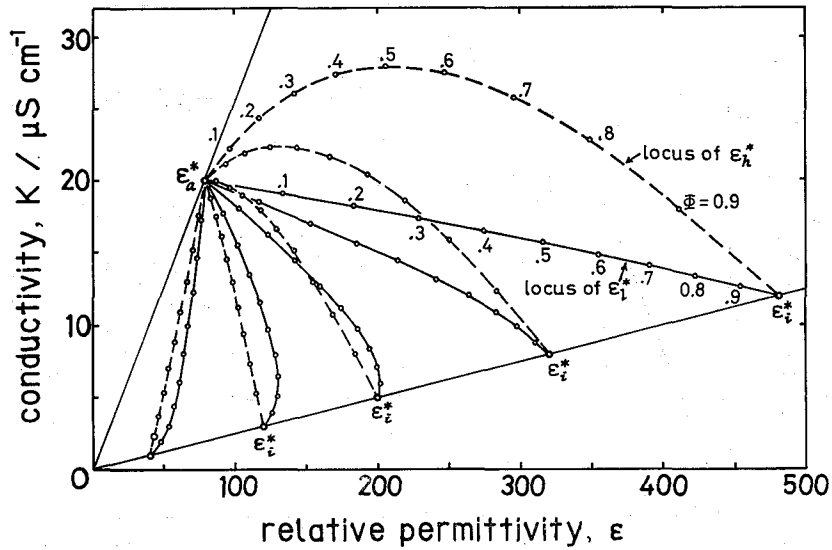


Fig. 2. Location on the  $\epsilon$ - $\kappa$  plane of  $\epsilon_i^*(\epsilon_i, \kappa_i)$  and  $\epsilon_h^*(\epsilon_h, \kappa_h)$  for concentrated disperse systems of spherical particles in a type of  $\kappa_i/\epsilon_i < \kappa_a/\epsilon_a$  (O/W). The solid curves show the loci of  $\epsilon_i^*$ , and the dashed curves denote the loci of  $\epsilon_h^*$  for varied concentrations  $\Phi$  under a fixed  $\epsilon_a^*(\epsilon_a, \kappa_a)$  and some of  $\epsilon_i^*(\epsilon_i, \kappa_i)$ .

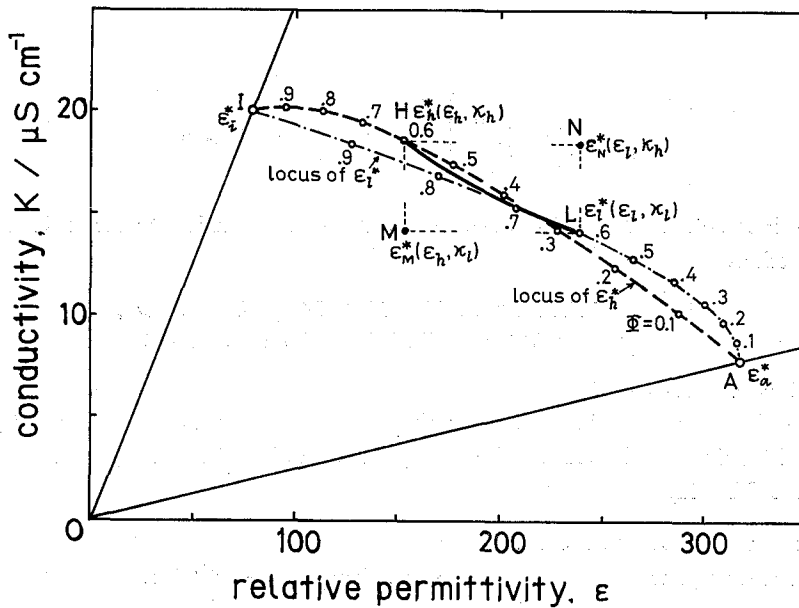


Fig. 3. Location of the  $\epsilon$ - $\kappa$  plane of  $\epsilon_i^*$ ,  $\epsilon_h^*$  and  $\epsilon^*(f)$ ,  $\epsilon_a=320$ ,  $\kappa_a=8 \mu S cm^{-1}$ ,  $\epsilon_i=80$ ,  $\kappa_i=20 \mu S cm^{-1}$ . The solid curve shows the locus of  $\epsilon^*(\epsilon(f), \kappa(f))$  for a fixed concentration  $\Phi=0.6$ , and the dotted broken curve denotes the locus of  $\epsilon_i^*$ , the dashed curve the locus of  $\epsilon_h^*$ .

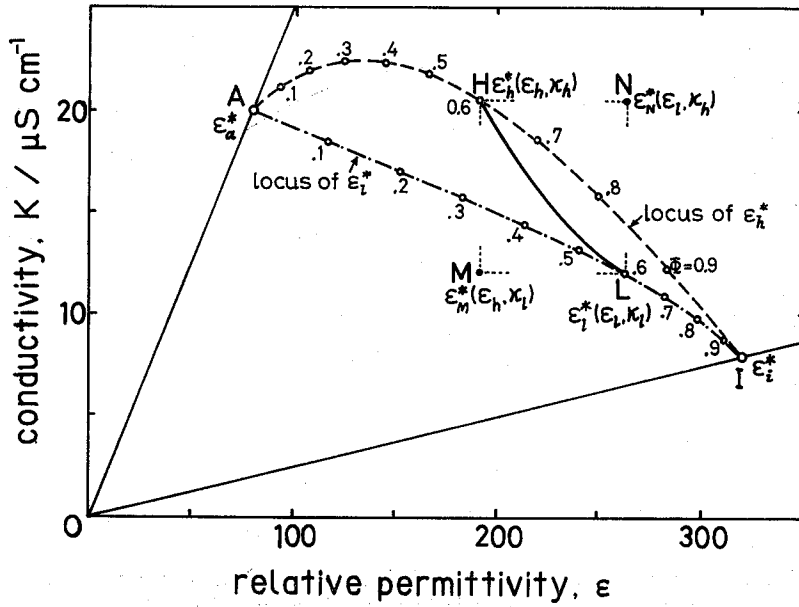


Fig. 4. Location on the  $\epsilon$ - $\kappa$  plane of  $\epsilon_i^*$ ,  $\epsilon_h^*$  and  $\epsilon^*(f)$ .  $\epsilon_a=80$ ,  $\kappa_a=20 \mu\text{S cm}^{-1}$ ,  $\epsilon_i=320$ ,  $\kappa_i=8 \mu\text{S cm}^{-1}$ .

The solid curve shows the locus of  $\epsilon^*(\epsilon(f), \kappa(f))$  for a fixed concentration  $\Phi=0.6$ , and the chain dotted broken curve denotes the locus of  $\epsilon_i^*$ , the dashed curve the locus of  $\epsilon_h^*$ .

and

$$\frac{\kappa_i}{\epsilon_i} < \frac{\kappa_l}{\epsilon_l} < \left\{ \frac{\kappa_l}{\epsilon_h} \right\} < \frac{\kappa_h}{\epsilon_h} < \frac{\kappa_a}{\epsilon_a} \text{ in Fig. 4.} \quad (19)$$

These inequalities 18 and 19 will be derived by mathematical formulas in the succeeding sections.

#### IV. RESTRICTION OF EXISTING DOMAINS OF $\kappa_a/\epsilon_a$ BY MEANS OF MATHEMATICAL FORMULAS

##### 4.1 Definition of Emulsion Type

It is well known that dielectric relaxations cease to be observable when  $\kappa_a/\epsilon_a = \kappa_i/\epsilon_i$ . From a viewpoint of dielectric relaxations, therefore, it is natural to define emulsion types as the following:

- |  |   |  |      |
|--|---|--|------|
| <ul style="list-style-type: none"> <li>a. water-in-oil (W/O) type emulsions</li> <li>b. homogeneous solutions</li> <li>c. oil-in-water (O/W) type emulsions</li> </ul> | } | according as $T \equiv \frac{\epsilon_a \kappa_i}{\kappa_a \epsilon_i} \geq 1$ . | (20) |
|--|---|--|------|

A new function  $S$  is further introduced as

$$S \equiv \frac{T}{D^3} = \frac{\epsilon_a \kappa_i}{\kappa_l \epsilon_i}. \quad (21)$$

#### 4.2 Existing Domain of $\kappa_l/\varepsilon_h$

For cases of physical significance,  $\varepsilon_h$  and  $\kappa_l$  given by Eqs. 5 and 8 are monotonic functions with respect to  $\Phi$ , being restricted as follows:

$$\text{either} \quad 0 < \varepsilon_a < \varepsilon_h < \varepsilon_i, \quad (22)$$

$$\text{or} \quad \varepsilon_a > \varepsilon_h > \varepsilon_i > 0, \quad (23)$$

and

$$\text{either} \quad 0 < \kappa_a < \kappa_l < \kappa_i, \quad (24)$$

$$\text{or} \quad \kappa_a > \kappa_l > \kappa_i > 0. \quad (25)$$

Therefore we have

$$0 < \frac{\varepsilon_h - \varepsilon_i}{\varepsilon_a - \varepsilon_i} < 1, \quad 0 < \frac{\varepsilon_a - \varepsilon_h}{\varepsilon_a - \varepsilon_i} < 1, \quad 0 < \frac{\varepsilon_h - \varepsilon_i}{\varepsilon_a - \varepsilon_h}, \quad (26)$$

and

$$0 < \frac{\kappa_l - \kappa_i}{\kappa_a - \kappa_i} < 1, \quad 0 < \frac{\kappa_a - \kappa_l}{\kappa_a - \kappa_i} < 1, \quad 0 < \frac{\kappa_l - \kappa_i}{\kappa_a - \kappa_l}. \quad (27)$$

From Eqs. 15, 5 and 26, we have

$$0 < C = \left( \frac{\varepsilon_h}{\varepsilon_a} \right)^{1/3} (1 - \Phi) = \frac{\varepsilon_h - \varepsilon_i}{\varepsilon_a - \varepsilon_i} < 1, \quad (28)$$

and

$$0 < 1 - C = \frac{\varepsilon_a - \varepsilon_h}{\varepsilon_a - \varepsilon_i} < 1. \quad (29)$$

From Eqs. 8, 14, 28 and 27, we have

$$0 < CD = \left( \frac{\kappa_l}{\kappa_a} \right)^{1/3} (1 - \Phi) = \frac{\kappa_l - \kappa_i}{\kappa_a - \kappa_i} < 1, \quad (30)$$

and

$$0 < 1 - CD = \frac{\kappa_a - \kappa_l}{\kappa_a - \kappa_i} < 1. \quad (31)$$

From Eq. 16, we have

$$\varepsilon_h - \varepsilon_i = \frac{(\varepsilon_a - \varepsilon_h)C}{1 - C} \quad \text{and} \quad \varepsilon_a - \varepsilon_i = \frac{\varepsilon_a - \varepsilon_h}{1 - C}. \quad (32)$$

From Eq. 17, we have

$$\kappa_l - \kappa_i = \frac{(\kappa_a - \kappa_l)CD}{1 - CD} \quad \text{and} \quad \kappa_a - \kappa_i = \frac{\kappa_a - \kappa_l}{1 - CD}. \quad (33)$$

For the later calculation, the following transformation of an equation is derived by use of Eqs. 28, 29 and 16.



$$\begin{aligned} \frac{\varepsilon_h(\kappa_l - \kappa_a CD)}{\kappa_l(\varepsilon_h - \varepsilon_a C)} &= 1 + \frac{\frac{\varepsilon_a}{\varepsilon_h} C - \frac{\kappa_a}{\kappa_l} CD}{1 - \frac{\varepsilon_a}{\varepsilon_h} C} \\ &= 1 + \frac{\varepsilon_a}{\varepsilon_i} \frac{(\varepsilon_h - \varepsilon_i)}{(\varepsilon_a - \varepsilon_h)} \frac{(D^2 - 1)}{D^2}. \end{aligned} \quad (34)$$

Therefore, we can derive the following relation by use of Eqs. 21 and 34.

$$S = \left[ 1 + \frac{C(D-1)}{1-CD} \right] \left[ 1 + \frac{\varepsilon_a}{\varepsilon_i} \frac{(\varepsilon_h - \varepsilon_i)}{(\varepsilon_a - \varepsilon_h)} \frac{(D+1)}{D^2} (D-1) \right]. \quad (35)$$

Taking Eqs. 20 and 21 into account, we have the following relations

$$S \cong 1, D \cong 1 \text{ according as } \begin{cases} T > 1, \text{ W/O type emulsion.} \\ T = 1, \text{ homogeneous solution.} \\ T < 1, \text{ O/W type emulsion.} \end{cases} \quad (36)$$

This relation 36 is transformed as

$$\frac{\kappa_i}{\varepsilon_i} \cong \frac{\kappa_l}{\varepsilon_h} \cong \frac{\kappa_a}{\varepsilon_a} \text{ according as } D \cong 1, \quad (37)$$

which are the part of relations 18 and 19.

#### 4.3 Restriction with $\kappa_l/\varepsilon_i$

From Eq. 6, we have

$$\frac{\varepsilon_l}{\kappa_l} = \frac{\varepsilon_a(2\kappa_a + \kappa_i)(\kappa_l - \kappa_i) + 3\varepsilon_i\kappa_a(\kappa_a - \kappa_l)}{(2\kappa_l + \kappa_i)(\kappa_a - \kappa_i)\kappa_a}. \quad (38)$$

By use of Eq. 38, we derive

$$\frac{\varepsilon_l}{\kappa_l} - \frac{\varepsilon_a}{\kappa_a} = \frac{3(\kappa_a - \kappa_l)}{(\kappa_a - \kappa_i)} \frac{\varepsilon_i\varepsilon_a}{(2\kappa_l + \kappa_i)\kappa_a} \left( \frac{\kappa_a}{\varepsilon_a} - \frac{\kappa_i}{\varepsilon_i} \right). \quad (39)$$

Therefore we have

$$\frac{\kappa_l}{\varepsilon_l} \cong \frac{\kappa_a}{\varepsilon_a} \text{ according as } D \cong 1. \quad (40)$$

#### 4.4 Restriction with $\kappa_h/\varepsilon_h$

From Eq. 7, we have

$$\frac{\kappa_h}{\varepsilon_h} = \frac{\kappa_a(2\varepsilon_a + \varepsilon_i)(\varepsilon_h - \varepsilon_i) + 3\varepsilon_i\varepsilon_a(\varepsilon_a - \varepsilon_h)}{(2\varepsilon_h + \varepsilon_i)(\varepsilon_a - \varepsilon_i)\varepsilon_a}. \quad (41)$$

By use of Eq. 41, we derive

$$\frac{\kappa_h}{\varepsilon_h} - \frac{\kappa_a}{\varepsilon_a} = \frac{3(\varepsilon_a - \varepsilon_h)}{(\varepsilon_a - \varepsilon_i)} \frac{\varepsilon_i}{(2\varepsilon_h + \varepsilon_i)} \left( \frac{\kappa_i}{\varepsilon_i} - \frac{\kappa_a}{\varepsilon_a} \right). \quad (42)$$

Therefore, we have

$$\frac{\kappa_h}{\varepsilon_h} \cong \frac{\kappa_a}{\varepsilon_a} \text{ according as } D \cong 1. \quad (43)$$

#### 4.5 Relation between $\kappa_i/\varepsilon_i$ and $\kappa_h/\varepsilon_h$

By use of Eq. 42, we have

$$\frac{\kappa_i}{\varepsilon_i} - \frac{\kappa_h}{\varepsilon_h} = \frac{(2\varepsilon_a + \varepsilon_i)}{(2\varepsilon_h + \varepsilon_i)} \frac{(\varepsilon_h - \varepsilon_i)}{(\varepsilon_a - \varepsilon_i)} \left( \frac{\kappa_i}{\varepsilon_i} - \frac{\kappa_a}{\varepsilon_a} \right). \quad (44)$$

Therefore, we have

$$\frac{\kappa_i}{\varepsilon_i} \cong \frac{\kappa_h}{\varepsilon_h} \text{ according as } D \cong 1. \quad (45)$$

#### 4.6 Relation between $\kappa_i/\varepsilon_i$ and $\kappa_l/\varepsilon_l$

By use of Eq. 39, we have

$$\frac{\varepsilon_i}{\kappa_i} - \frac{\varepsilon_l}{\kappa_l} = \frac{(2\kappa_a + \kappa_i)}{(2\kappa_l + \kappa_i)} \frac{(\kappa_l - \kappa_i)}{(\kappa_a - \kappa_i)} \left( \frac{\varepsilon_i}{\kappa_i} - \frac{\varepsilon_a}{\kappa_a} \right). \quad (46)$$

Therefore, we have

$$\frac{\kappa_i}{\varepsilon_i} \cong \frac{\kappa_l}{\varepsilon_l} \text{ according as } D \cong 1. \quad (47)$$

#### 4.7 Summary of Existing Domains

From Eqs. 37, 40, 43, 45 and 47, the relative magnitude of  $\kappa_a/\varepsilon_a$ ,  $\kappa_l/\varepsilon_l$ ,  $\kappa_i/\varepsilon_i$ ,  $\kappa_h/\varepsilon_h$  and  $\kappa_i/\varepsilon_i$  can be summarized as follows:

For W/O type,

$$\frac{\kappa_a}{\varepsilon_a} < \frac{\kappa_l}{\varepsilon_l} < \frac{\kappa_i}{\varepsilon_i} < \frac{\kappa_h}{\varepsilon_h} < \frac{\kappa_i}{\varepsilon_i} \text{ when } D > 1, T > 1, S > 1. \quad (48)$$

For homogeneous solutions,

$$\frac{\kappa_a}{\varepsilon_a} = \frac{\kappa_l}{\varepsilon_l} = \frac{\kappa_i}{\varepsilon_i} = \frac{\kappa_h}{\varepsilon_h} = \frac{\kappa_i}{\varepsilon_i} \text{ when } D = 1, T = 1, S = 1. \quad (49)$$

For O/W type,

$$\frac{\kappa_i}{\varepsilon_i} < \frac{\kappa_l}{\varepsilon_l} < \frac{\kappa_i}{\varepsilon_i} < \frac{\kappa_h}{\varepsilon_h} < \frac{\kappa_a}{\varepsilon_a} \text{ when } D < 1, T < 1, S < 1. \quad (50)$$

These relations 48, 49 and 50 which were derived analytically are in conformity with inequality relations 18 and 19 which were understood graphically in Figs. 3 and 4.

The existing domains of  $\kappa_a/\varepsilon_a$  and  $\kappa_i/\varepsilon_i$  are illustrated in Fig. 5 for the restrictions by Eqs. 37, 40, 43, 45 and 47 and for the resultant restrictions. According to the figure, the existing domain of  $\kappa_a/\varepsilon_a$  is either  $\kappa_a/\varepsilon_a < \kappa_l/\varepsilon_l$  or  $\kappa_h/\varepsilon_h < \kappa_a/\varepsilon_a$  according as  $D > 1$  or  $D < 1$ .

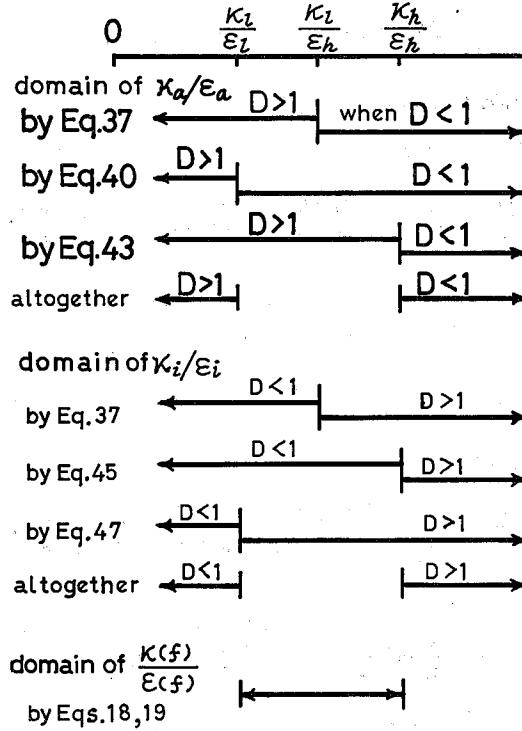


Fig. 5. Existing domains of  $\kappa_a/\epsilon_a$ ,  $\kappa_i/\epsilon_i$  and  $\kappa(f)/\epsilon(f)$ .

## V. SEARCHING DIRECTION OF $\kappa_a/\epsilon_a$ TO FIND THE CORRECT ROOT OF $J(\kappa_a)=0$

Provided that  $\epsilon_h$ ,  $\kappa_l$ ,  $\epsilon_a$  and an approximate value of  $\kappa_a$  are known by experiments, a quantity  $D^3$  is readily calculated by the relation

$$\frac{\epsilon_a \kappa_l}{\kappa_a \epsilon_h} = D^3, \quad (51)$$

which is identical with Eq. 14. Next, a root  $\kappa_a$  of  $J(\kappa_a)=0$  given by Eq. 9 is searched by varying the value of  $\kappa_a$ . In this instance, the value of  $\kappa_a$  starts from  $\epsilon_a \kappa_l / \epsilon_l$  and towards the smaller values of  $\kappa_a$  when  $D^3 > 1$ . When  $D^3 < 1$ , then the searching for  $\kappa_a$  should be proceeded from  $\epsilon_a \kappa_h / \epsilon_h$  and towards the larger values of  $\kappa_a$ .

As regards the criterion of the searching direction, either  $T \geq 1$  or  $S \geq 1$  can be adopted in place of  $D^3 \geq 1$ .

## VI. SOME NUMERICAL EXAMPLES

In order to examine the applicability of the improved evaluation proposed above, five numerical examples are shown.

First of all, a set of phase parameters  $\epsilon_a$ ,  $\epsilon_i$ ,  $\kappa_a$ ,  $\kappa_i$  and  $\phi$  was assumed. Next, the dielectric parameters  $\epsilon_h$ ,  $\kappa_l$ ,  $\epsilon_l$  and  $\kappa_h$  were calculated from Eqs. 5, 8, 6 and 7 by use of the values of  $\epsilon_a$ ,  $\epsilon_i$ ,  $\kappa_a$ ,  $\kappa_i$  and  $\phi$  which were assumed above. Those values for five

Table I. Numerical Values of the Parameters in Some Examples to Test the Improved Method of Computer Searching for the Correct Root of  $J(\kappa_a)=0$  Given by Eq. 9.

	Example 1	Example 2	Example 3	Example 4	Example 5
Figure shown	Fig. 6	Fig. 7	Fig. 8	Fig. 9	Fig. 10
Emulsion type	O/W	O/W	W/O	W/O	W/O
$\epsilon_a$	2	80	80	2	80
$\epsilon_i$	80	2	2	80	2
$\kappa_a/\mu\text{S cm}^{-1}$	30	100	1	1	1
$\kappa_i/\mu\text{S cm}^{-1}$	1	1	0.3	1000	30
$\Phi$	0.8	0.8	0.8	0.8	0.8
$\epsilon_i$	95.86	9.774	11.78	144.9	325.7
$\epsilon_h$	38.27	9.729	9.729	38.27	9.729
$\kappa_i/\mu\text{S cm}^{-1}$	3.951	10.27	0.4035	93.42	15.53
$\kappa_h/\mu\text{S cm}^{-1}$	165.1	10.32	0.4586	346.6	36.85
$D^3 = \frac{\epsilon_a \kappa_i}{\kappa_a \epsilon_h}$	0.006882	0.8447	3.318	4.882	127.7
$\epsilon_a \frac{\kappa_i}{\epsilon_i} / \mu\text{S cm}^{-1}$	0.08243	84.08	2.741	1.289	3.815
$\epsilon_a \frac{\kappa_i}{\epsilon_h} / \mu\text{S cm}^{-1}$	0.2065	84.48	3.318	4.882	127.7
$\epsilon_a \frac{\kappa_h}{\epsilon_h} / \mu\text{S cm}^{-1}$	8.630	84.88	3.771	18.11	303.0
$\epsilon_a \frac{\kappa_i}{\epsilon_i} / \mu\text{S cm}^{-1}$	0.025	40.0	12.0	25.0	1200

examples are listed in Table I together with the values of  $D^3$ ,  $\epsilon_a \kappa_i / \epsilon_i$ ,  $\epsilon_a \kappa_i / \epsilon_h$ ,  $\epsilon_a \kappa_h / \epsilon_h$  and  $\epsilon_a \kappa_i / \epsilon_i$ .

The curve of  $J(\kappa_a)$  which is given by Eq. 9 as a function of  $\kappa_a$  is depicted in Figs. 6, 7, 8, 9 and 10. In these figures, the arrows  $\rightarrow$  show the present improved searching direction (abbreviated to ISD) for the correct root (CR) of  $J(\kappa_a)=0$  following the new evaluation proposed above.

In the previous paper, a scheme of the primitive searching direction (PSD) for the correct root of  $J(\kappa_a)=0$  was suggested as follows:

- (i) If  $\kappa_a < \kappa_l < \kappa_i$ , then the value of  $\kappa_a$  starts from  $\kappa_l$  for smaller values of  $\kappa_a$ .
- (ii) If  $\kappa_a > \kappa_l > \kappa_i$ , then the value of  $\kappa_a$  starts from  $\kappa_l$  for larger values of  $\kappa_a$ .

These primitive searching directions are shown with double arrows  $\Rightarrow$  in the figures.

#### Example 1 (Fig. 6)

Both the improved searching direction (ISD)  $\rightarrow$  and the primitive searching direction (PSD)  $\Rightarrow$  attain the correct root (CR) of  $J(\kappa_a)=0$  straightway.

#### Example 2 (Fig. 7)

The primitive searching starts from  $\kappa_l$  for larger values of  $\kappa_a$  (the right direction), and meets with an unidentified false root (UFR) and a point for  $D=1$ . Afterwards the searching attains the correct root.

The improved searching starts from  $\epsilon_a \kappa_h / \epsilon_h$  for larger values of  $\kappa_a$ , and attains the correct root straightway.

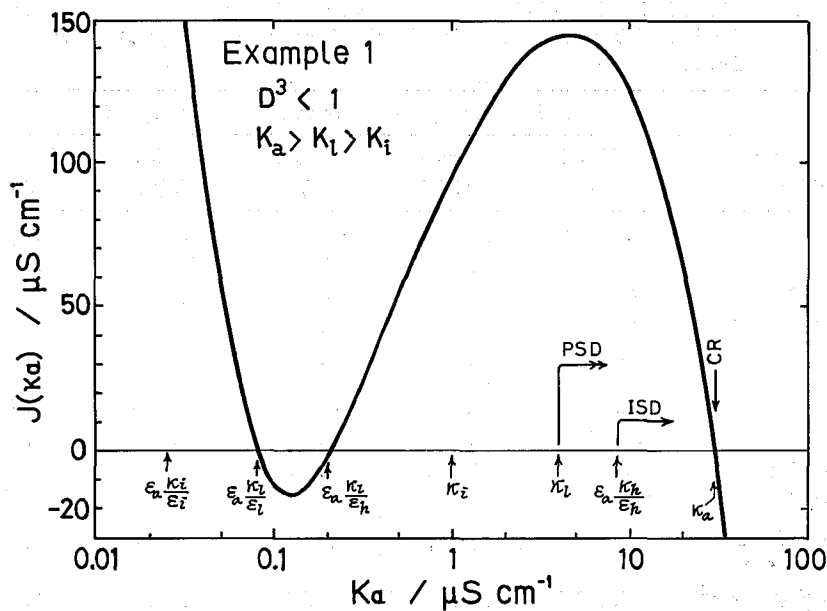


Fig. 6. The curve of the function  $J(\kappa_a)$  given by Eq. 9 against  $\kappa_a$  for Example 1. In the figure, PSD with a double arrow  $\Rightarrow$  means the primitive searching direction, and ISD with an arrow  $\rightarrow$  denotes the improved searching direction. An abbreviation CR shows the location of the correct root of  $J(\kappa_a) = 0$ . The numerical values of the parameters used are listed in Table I.

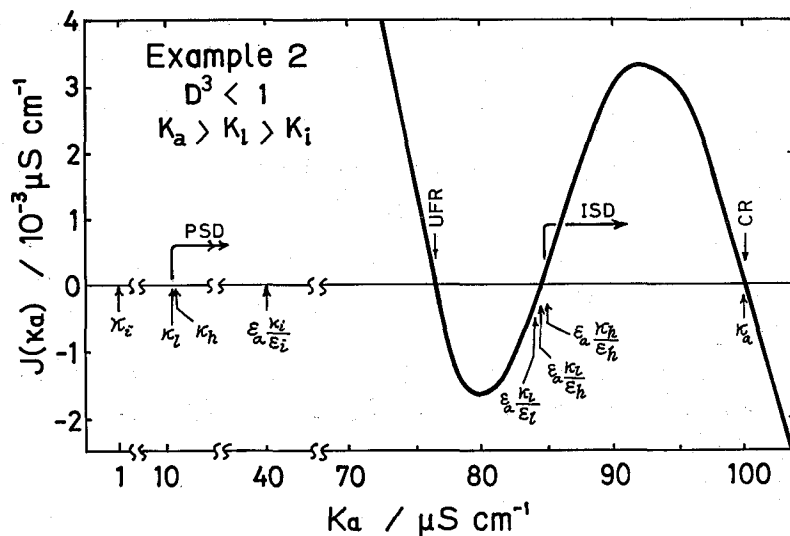


Fig. 7. The curve of the function  $J(\kappa_a)$  given by Eq. 9 against  $\kappa_a$  for Example 2.

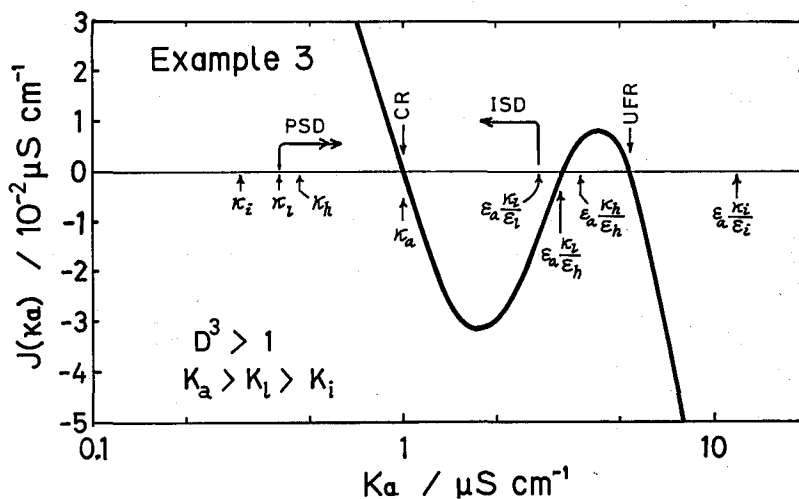


Fig. 8. The curve of the function  $J(\kappa_a)$  given by Eq. 9 against  $\kappa_a$  for Example 3.

#### Example 3 (Fig. 8)

The primitive searching starts from  $\kappa_l$  for the right direction, attaining the correct root.

The improved searching starts from  $\varepsilon_a \kappa_l / \varepsilon_l$  for the left direction, attaining the correct root.

#### Example 4 (Fig. 9)

The primitive searching starts from  $\kappa_l$  for the left direction, and meets with an unidentified false root (UFR) and a point for  $D=1$ . This unidentified false root gives a set of plausible values:  $\phi=0.6602$ ,  $\varepsilon_i=400.7$  and  $\kappa_i=176.0 \mu\text{S cm}^{-1}$ . The farther searching attains the correct root.

The improved searching starts from  $\varepsilon_a \kappa_l / \varepsilon_l$  for the left direction, and attains the correct root straightway.

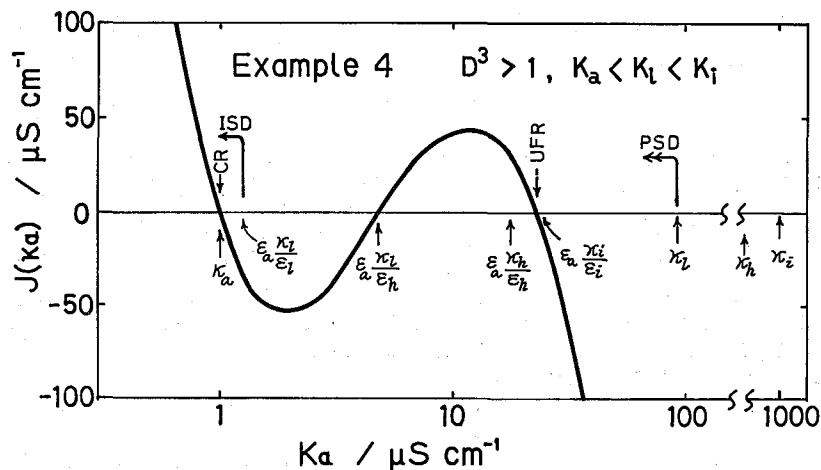


Fig. 9. The curve of the function  $J(\kappa_a)$  given by Eq. 9 against  $\kappa_a$  for Example 4.

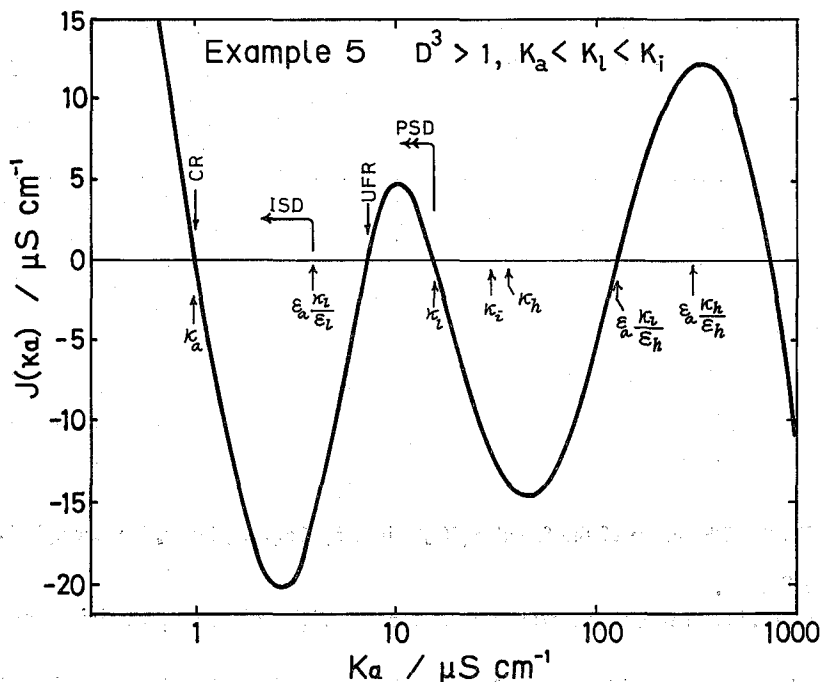


Fig. 10. The curve of the function  $J(\kappa_a)$  given by Eq. 9 against  $\kappa_a$  for Example 5.

#### Example 5 (Fig. 10)

The primitive searching starts from  $\kappa_l$  for the left direction, and meets with an unidentified false root (UFR). This unidentified false root gives a set of unrealistic values:  $\phi=0.129$ ,  $\varepsilon_i=-43.6$  and  $\kappa_i=-56.2 \mu\text{S cm}^{-1}$ . Afterwards the searching attains the correct root.

The improved searching starts from  $\varepsilon_a \kappa_l / \varepsilon_i$  for the left direction, and attains the correct root straightway.

It is concluded that the improved searching always attains the correct root without meeting with the false roots in contrast with the primitive searching scheme which often meets with false roots.

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